On the Magnetic Instability of Fe_xN_{1-x} Pseudomorphic Thin Films Exhibiting The Invar Effect

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INTRODUCTION

At a critical concentration of around 65% Fe, bulk FeNi alloys exhibit the "Invar effect", a sudden arresting of the Wigner-Seitz cell volume and a zero expansion coefficient. Simultaneously, the crystal structure changes from face-centered cubic (fcc) to body-centered cubic while the ferromagnetic Curie temperature goes to zero [1]. The 'martensitic' structural transformation can be arrested in ultrathin alloys films grown epitaxially on a Cu(001) substrate [2]. The electronic mechanism(s) underpinning this instability remain obscure, despite much experimental and theoretical effort. Theoretical work [3] predicts that the average magnetic moment in these metastable structures collapses to zero when the Fe content increases beyond the critical "Invar" composition (65%), accompanied by a sudden reduction of the Wigner-Seitz cell volume of 9%. Experimental work [4] shows a lattice expansion increasing linearly up to 3% at 65% Fe content followed by a sudden relaxation of 2% with increasing Fe content.

The initial volume increase is associated with increasing magnetization/magnetic moment & spin alignment in the Ni-rich alloys and an increasing Curie temperature T_C. The small coefficient of thermal expansion observed over a wide temperature range below T_C is due to "spontaneous volume magnetostriction effect" opposing the usual Grüneisen change in lattice contraction on cooling below T_C [1]. As the alloy is cooled below T_C (or a strong external magnetic field is applied), an increasing alignment of the magnetic moments causes the nearest-neighbor spins to push apart producing an internal pressure which expands the lattice. With increasing Fe content, this effect increases due to the increasing number of Fe nearest neighbors with the larger atomic magnetic moments. Eventually, a critical limit is reached (~65% Fe), when a magnetic/lattice volume instability develops. The coexistence of a "High-spin" ferromagnetic phase slowly converting to a ferromagnetically disordered or antiferromagnetically ordered phase has been argued on the basis of Mössbauer spectroscopy measurements [5].

The evolution of the elemental magnetic moments was tracked using linearly-polarized x-ray magnetic dichroism angle-dependent (XMLDAD) 3p core-level photoemission [6]. Spin polarized photoemission studies record a sudden decrease in the "mean-field" exchange splitting of the d-states with increasing Fe content through the critical "Invar transition". Angle-resolved photoemission imaging of states at the Fermi level [7] reveal a much smaller splitting of the sp-states, which also tracks the changing magnetization with changing composition. Spectral lineshapes reveal a decreased lifetime (i.e. decreased mean-free path for scattering) of the minority spin-polarized sp-states, in agreement with reported similar measurements on permalloy [8].

RESULTS AND DISCUSSION

A plot of the change in the asymmetry amplitude, for both elements in the FeNi alloy measured with XMLDAD, being a measure of the expectation value of the atomic magnetic moment <μ>, is shown plotted as a function of composition, fig. 1 (left panel). We observe that both the Ni and Fe signals track a similar profile with changing composition. In the Ni-rich alloys, both signal increase linearly up to 65% that on the Fe showing the larger increase. Above 65% Fe content, both signals show a sharp decrease. The observed asymmetry amplitudes, suggest that a high-spin moment develops on the Fe with increasing Fe content that increases overall magnetization, which then increases the polarization of the valence states surrounding the Ni atomic cores. The Ni thus develops a component that tracks the developing magnetization. Above 65% Fe-content, the high-spin moment on the Fe appears to collapse to a ”low-spin value”, causing the overall magnetization density to be lowered, which is sensed by the reduced polarization of the valence states on the Ni. A plot of the variation of a ‘stoichiometric average moment’ $xA(Fe_x)+(1-x)A(Ni_{1-x})$ is shown in fig. 1 (right panel).. The behavior is very similar to that reported for the variation of the saturated moment normalized to the volume of similar fcc films on Cu(100) & Cu(111) and measured with SQUID magnetometry [5]. The solid line is the behavior reported for FeNi alloys from neutron scattering measurements [1]. We note that the Ni-rich phase extrapolates to a value around μ=2.5-3.0μB, a value predicted theoretically for the “high-moment” metastable fcc phase [3]. The above ‘mean magnetic moment’ variation, normalizing the Ni asymmetry amplitude to be equivalent to the magnetic moment of metallic Ni is tracking closely the Slater-Pauling curve, the moment increasing linearly with increasing number of holes per atom in the valence electronic states. Above 65% Fe content, the average moment shows a sharp decline into a “low-spin” magnitude state, which could be the result of a collapse of the spin moment on the Fe atoms and/or a sudden decrease in magnetization due to a non collinear rearrangement of spins.

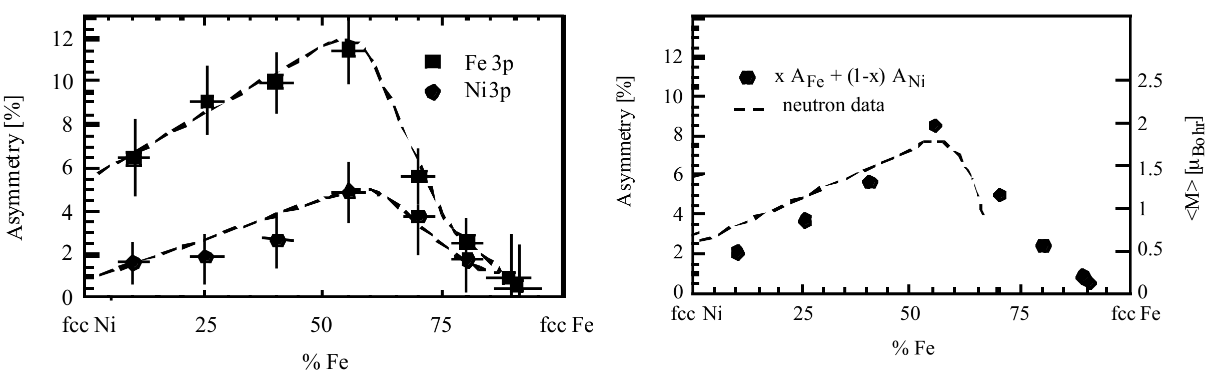

Figure 1. Left panel: Change in dichroism amplitude, A, as a function of FeNi alloy composition. Right panel: The variation of the ‘stoichiometrically-weighted’ dichroism signal amplitude (see text) with changing FeNi alloy composition. The dashed curve is the behavior observed in bulk FeNi alloys by neutron scattering. The right hand scale is determined from neutron and SQUID magnetometry data [1,5]

Angle-resolved photoemission measurements of the sp-states, away from the regions of emerging minority d-states, along the <110> (Σ) symmetry direction, resolves the sp exchange splitting in reciprocal space, fig. 2 (left panel). We observe that the spectral width of the minority-spin band of the sp-states is broader than that of the majority-spin sp-band.

This has been reported in similar measurements on permalloy, and is indicative of a shorter lifetime due to increased scattering and a shorter mean-free-path for the minority spin electrons. We also note that the lifetime broadening of the minority-spin sp-states increases significantly in the Fe rich alloys.

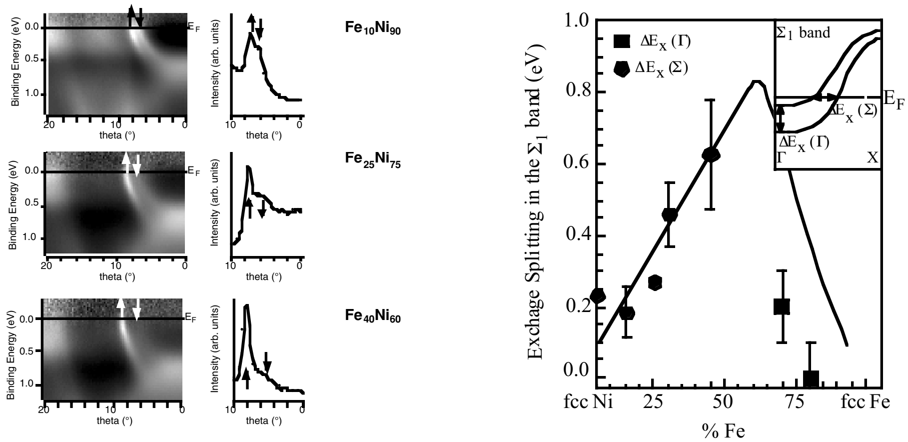

Figure 1. Left panel: Photoemission images of dispersion of exchange-split majority (spin-up) and minority (spin-down) sp-states and line scans along <110> (Σ) direction at E_F shows the different lifetimes of these states and their evolution with increasing Fe content. Right panel: Variation of the exchange-splitting of the valence band d-states and sp-states. The mean magnetic moment behavior is superimposed (full curve).

Figure 2 (right panel). shows the measured exchange splitting of the sp-states as well as the spin-resolved measured exchange splitting of the d-states. Both, sp-states exchange splitting and d-states exchange splitting, track the behavior of the x-ray core-level photoemission dichroism.
This is to be expected on the basis of the overall magnetic energy being the sum of a 'local moment' energy on the 'atom(s) and a 'mean-field' exchange energy rising from the spin polarization of the itinerant valence states [9].

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